Elastic constant dishomogeneity and Q^2 dependence of the broadening of the dynamical structure factor in disordered systems

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We propose an explanation for the quadratic dependence on the momentum Q, of the broadening of the acoustic excitation peak recently found in the study of the dynamic structure factor of many real and simulated glasses. We ascribe the observed Q^2 law to the spatial fluctuations of the local wavelength of the collective vibrational modes, in turn produced by the dishomegeneity of the inter-particle elastic constants. This explanation is analitically shown to hold for 1-dimensional disordered chains and satisfatorily numerically tested in both 1 and 3 dimensions.

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The study of the nature of collective atomic excitations in disordered solids at wavelengths λ approaching the interparticle separation a, $a/\lambda \approx 0.1 - 0.5$, has received renewed interest in the last few years thanks to new experimental tools and to improved numerical techniques. On the experimental side, the dynamics of the collective excitations is often investigated via the dynamic structure factor $S(Q,\omega)$, i. e. the space-time Fourier transform of the particle density correlation function. The study of $S(Q,\omega)$ in the region of mesoscopic exchanged momentum ($Q = 1 - 10$ nm^{-1}) has become recently possible in disordered systems thanks to the development of the Inelastic X-ray Scattering (IXS) technique [1], and many glasses [2] and liquids [3] have been studied with this technique. Although specific *quantitative* differences exist among different systems, all the investigated glasses show some common features that can be summarized as follows: *i)* there exist propagating acoustic-like excitations up to $Qa \approx 3$; *ii)* the slope of the $Q - \omega$ dispersion relation in the $Q \to 0$ limit extrapolates to the macroscopic sound velocity; *iii)* the broadening of the excitation peaks in $S(Q,\omega)$ follows a Q^2 law. These general features of $S(Q,\omega)$ have been confirmed by numerical calculations in different glasses, using both standard Molecular Dynamics (MD) simulations [4], and the Normal Mode Analysis (NMA) in the harmonic approximation [5].

The Q^2 dependence of the excitations broadening has not yet received a theoretical explanation. Such dependence is the same as predicted by hydrodynamics, but this coincidence is only accidental. Indeed, in the experiments the broadening is found to be temperature independent and the Q^2 law is numerically found also in *harmonic* glass models; these results indicate that the origin of this behavior should be found in *structural* rather than *dynamical* properties, i. e. it should be associated to the atomic disorder in the glass and not to dissipative phenomena like anharmonicity or relaxation processes. For this reasons, we concentrate on harmonic systems, leaving apart all the difficulties due to dynamical processes like, for example, anharmonicity.

In this Letter we suggest that the observed broadening is due to the spatial fluctuations of the elastic constants. Our explanation is based on analytical and numerical results for the simple case of a 1-dimensional (1-d) harmonic disordered system (linear chain of like mass joined by random springs), and then it is generalized to 3 dimensions.

The dynamics of 1-d disordered lattices has been thoroughly investigated in the past,and a recent overview on this subject can be found in [6]. In principle, 1-d systems often show very peculiar characteristics; more specifically it is well known [7] that, at variance with d>1 cases, in a (infinite) disordered linear chain all vibrational modes are localized and localization by itself contributes to the broadening of the inelastic peaks of $S(Q,\omega)$. However, as we will show, this effect can be disentangled. Our model consists of an linear chain of N particles ($i = 1..N$) of mass M placed a distance a apart from each other ($x_i = ia$), and joined by next-neighbour springs (K_i) randomly chosen from a flat distribution ($\mathcal{P}(K)$) with extrema $K \pm \Delta K$ (mean $\mu_k = K$ and standard deviation $\sigma_k = \Delta K/\sqrt{3}$). The dynamics of the system can be expressed in terms of its eigenvalues (ω_p) and eigenvectors ($e_p(i)$), $p = 1...N$ being the mode label. The classical dynamic structure factor is expressed as:

$$S(Q,\omega) = \frac{K_B T}{M}\frac{Q^2}{\omega^2}E(Q,\omega) \tag{1}$$

with

$$E(Q,\omega) = \sum_p |\tilde{e}_p(Q)|^2 \delta(\omega - \omega_p), \tag{2}$$

$$\tilde{e}_p(Q) = N^{-1/2}\sum_i \exp(iQx_i)e_p(i).$$

The eigenvectors and eigenvalues are the solutions of the secolar problem, $\sum_j D_{ij}e_p(j) = \omega_p^2 e_p(i)$ where D_{ij} is the dynamical matrix. The basic properties of $S(Q,\omega)$ are cointained in $E(Q,\omega)$, the squared space Fourier

transform of the eigenvectors of modes with frequency ω_p "close" to ω. A typical eigenvector obtained from the diagonalization of the dynamical matrix is reported in Fig. 1. As is well known (see for example [8]), there exists a rather well defined wavelength but, as compared with the sinus function expected in the case of an ordered system, three main differences can be noted: *i)* the peak height is not constant, i. e. there exists an envelope which is localized in space (see inset a); *ii)* the wavelength λ (i. e. the distance between two next nearest nodes) is not constant, but is rather a space fluctuating quantity; its statistical distribution is reported in the inset b) of Fig. 1; finally, *iii)* by analysing in detail the eigenvector between two successive nodes (see insets c) and d), some deviations from the simple sinus law can be evidenced. In principle, all these three characteristics could contribute to broade the inelastic peaks in $S(Q,\omega)$. In the following we will show that the broadening is mainly due to fluctuations of the local wavelength and (in 1-d systems) to localization, while *iii)* has minor consequences, and its signature can only be found in the low-ω tail of $S(Q,\omega)$.

Let us first focus on *ii)*, i. e. on the effect of spatial wavelength fluctuations. To show how these fluctuations by themselves can produce the Q^2-broadening, we build up a model eigenmode which possess only characteristic *ii)*, that is a sequence of alternating positive and negative semiperiods of a sinus function of slightly different wavelength. If h_m ($m = 1, 2...$) are the positions of the nodes, we define the local wavelengths $\lambda_m = h_m - h_{m-2}$ and compute the Fourier transform of the model eigenvectors, i.e. $\tilde{e}(Q) \propto \sum_m \int_{h_{m-2}}^{h_m} dx\ \exp(iQx)\ \ \sin(2\pi/\lambda_m(x - h_{m-2}))$. This is then averaged over different realizations of the disordered chain, assuming a gaussian distribution of wavelengths centered at λ and with variance σ_λ^2. The calculation of $E(Q) = N^{-1}\langle|\tilde{e}(Q)|^2\rangle$ is straightforward althought long; it produces an $E(Q)$ that near the peak, located at $Q = 2\pi/\lambda$, has a quasi-lorentzian shape with half width at half maximum (HWHM) $\Gamma_{_F}^{(Q)}$ given by:

$$\frac{\Gamma_{_F}^{(Q)}}{Q} = \pi\frac{\sigma_\lambda^2}{\lambda^2} \tag{3}$$

The successive step is to find a relationship between σ_λ and the characteristics of disorder. In each segment of length λ_m, containing n_m springs, we define a "local" sound velocity, $v_m^{loc} = a\sqrt{K_m^{eff}/M}$, where K_m^{eff} is the effective elastic constant obtained by averaging the individual spring constants K_i that are found between h_{m-2} and h_m: $(K_m^{eff})^{-1} = \sum_{i\in m} P_i K_i^{-1} / \sum_{i\in m} P_i$. Here the P_i's are weights that take into account the fact that springs near the nodes are more effective (highly stretched) than those near the antinodes (not stretched) in determining K_m^{eff}. The validity of the concept of "local velocity" can be tested numerically. In fact, coming back to the eigenvectors obtained by the solution of the secular problem for the disordered linear chain, the actual wavelength of the m-th half-period, λ_m, can be compared with that obtained from the local sound velocity, i. e. from the relation $\lambda_m^{loc} = 2\pi v_m^{loc}/\omega$. As an example, in Fig. 2 we show the correlation between λ and λ^{loc} obtained from the analysis of the eigenvectors of the modes at $\omega/\sqrt{K/M} \approx 0.031$ of 1,000 disordered chains with $\Delta K/K$=0.3, for different choices of the weights P_j. As can be seen, the correlations are satisfactory, indicating the validity of the assumptions that *i)* a local sound velocity exists, and *ii)* it is determined by a local spring constant averaged over the wavelength. Moreover, we observe that the highest correlation is found for weights proportional to the square of the local strain. From the previous relations it is easily deduced that:

$$\frac{\sigma_\lambda}{\lambda} = \frac{\sigma_v}{v} = \frac{1}{2}\frac{\sigma_{K^{eff}}}{K^{eff}} = \frac{1}{2}\sqrt{\frac{\alpha}{n}}\frac{\sigma_k}{\mu_k} \tag{4}$$

where $n = \langle n_m\rangle = \lambda/a$ and $\alpha = \langle P^2\rangle/\langle P\rangle^2$. In the following we will choose P proportional to the square of the strain and, therefore, $\alpha = 3/2$. By substituting Eq. 4 into 3, we obtain:

$$\Gamma_{_F}^{(Q)} = a\frac{\sigma_k^2}{\mu_k^2}\frac{\alpha}{8}Q^2 \tag{5}$$

which reproduces the experimental Q^2 behavior. Summing up, we have shown here that the spatial elastic constant fluctuations by themselves produce a broadened $E(Q,\omega)$ which is quasi-lorentzian in shape with HWHM $\Gamma_{_F}^{(Q)} \propto Q^2$.

In order to check the validity of Eq. 5 we performed a numerical calculation of $E(Q,\omega)$. At selected values of ω, and for different values of ΔK, we calculated the eigenvectors of 50 different realizations of a disordered chain composed of 20,000 atoms using the Dyson-Schmidt (DS) method [9]. Using Eq. 2 the functions $E(Q,\omega)$ are then calculted as a function of Q; representative examples are reported in Fig. 3 for the indicated values of ω and $\Delta K/K = 0.6$. The $E(Q,\omega)$ have been fitted to a lorentzian lineshape. The derived HWHM Γ are reported in Fig. 4 together with our prediction (Eq. 5), which reproduces correctly the Q^2 behavior but is about a factor 2 too small. This discrepancy is due to our neglecting of localization effects which, as mentioned before, are certainly effective in 1-d systems. In these systems the eigenvectors have an exponential envelope in the tails, whith a decay length L, and this produces a contribution to the linewidth of $E(Q,\omega)$ vs Q given by $\Gamma_{_L}^{(Q)} = 1/L$ [6]. Since localization also gives rise to a lorentzian lineshape, we expect a total broadening given by $\Gamma_{_{Tot}}^{(Q)} = \Gamma_{_L}^{(Q)} + \Gamma_{_F}^{(Q)}$. The values of L have been numerically computed by fitting the tails of the eigenvectors produced by the DS technique; as expected [6] we found $L \propto \omega^{-2}$ and, therefore, also in this case $\Gamma_{_L}^{(Q)} \propto Q^2$. The

full line in Fig. 4 represents the total HWHM $\Gamma^{(Q)}_{Tot}$: it is in quantitative agreement with the widths of $E(Q,\omega)$ determined numerically.

We stress that the effect of localization is peculiar to 1-d systems and is not relevant to 3-d disordered systems [5]. Therefore, we expect that in 3-d the wavelength fluctuations induced by the spatial flutuations in the local elastic constant account for most of the broadening observed in experiments. Since usually experiments probe $S(Q,\omega)$ as a function of ω at fixed Q, we rewrite Eq. 5 as

$$\Gamma^{(\omega)}_{F} = v\Gamma^{(Q)}_{F} = a\, v\frac{\sigma_k^2}{\mu_k^2}\frac{\alpha}{8}Q^2 \qquad (6)$$

where v is the sound velocity. In order to verify whether this equation, derived for 1-d, is still valid for 3-d systems we determined numerically the broadening of $S(Q,\omega)$ for a simple model glass consisting of 32,000 Ar atoms interacting via the 12-6 Lennard-Jones (LJ) potential ($\sigma = 3.405$ $\mathring{A}$, $\epsilon/K_B = 125.2$ K); for further details on this simulation see [10]. In the inset of Fig. 5 we report the $\Gamma^{(\omega)}$ values measured from a lorentzian fit to the numerically calculated $S(Q,\omega)$ (which are also shown in Fig. 5), compared with the prediction of Eq. 6 (full line). The values of σ_k, μ_k and a, needed to calculate $\Gamma^{(\omega)}_{F}$ from Eq. 6, were determined by averaging the interatomic distances and the second derivatives of the LJ interatomic potential [11]. The good quantitative agreement between the numerical and the predicted values of $\Gamma^{(\omega)}$ gives us confidence on the applicability of Eq. 6 to real glasses, and therefore on the prevalence of the elastic-constant fluctuation mechanism in determining the width of inelastic peaks in the $S(Q,\omega)$.

We emphasize that the mechanism proposed for the origin of the broadening of the Brillouin peaks in $S(Q,\omega)$ is completely different from the situation where a mode with well defined wavelength exists and its scattering is produced by isolated point defects (Rayleigh scattering), which would result in a Q^4 law.

In conclusion, we propose a plausible explanation for the ubiquitous Q^2 law found for the broadening of the Brillouin peaks in disordered systems. According to the present model this broadening is a consequence of the spatial fluctuation of the "effective" (i. e. averaged over one wavelength) elastic constant. This fluctuation becomes smaller and smaller as the wavelength (i. e., as the number of involved springs) increases and in particular, from basic statistics it is $\propto \lambda^{-1/2}$. This result, together with the existence of a "local velocity", leads to a Q^2 dependence of the Brillouin linewidth in harmonic disordered systems at all Q's.

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[11] The quantities a, μ_k, and σ_k, entering Eq. 6, were determined by the equations:

$$\langle O\rangle = \left[\int_o^{r_o} dr\; 4\pi\; r^2\; g(r)\; O(r)\right]\left[\int_o^{r_o} dr\; 4\pi\; r^2\; g(r)\right]^{-1}$$

where $O(r) = r$ for a, $O(r) = V''_{LJ}(r)$ for μ_k and $O(r) = (V''_{LJ}(r) - \mu_k)^2$ for σ_k^2. Here $V_{LJ}(r)$ is the LJ potential function, $g(r)$ is the pair distribution function of the system and r_o a cut-off radius that takes into account that in the theoretical derivation of Eq. 6 we only considered nearest neighbours interaction. The results only slightly depend on the choice of r_o, and we fixed it at half way between the first and second peak in the $g(r)$.

FIGURE CAPTIONS

FIG. 1 Portion of the eigenvector of the mode at $\omega/\sqrt{K/M} = 0.571$ of a chain with N=1,000 particles and $\Delta K/K = 0.6$ reported as a function of the particle coordinate. In the insets the three most relevant features of the eigenvectors are emphasized: *i)* Localization (the envelope of the whole eigenvector is reported in inset a); *ii)* Wavelength fluctuations (the distribution of λ_m calculated from 100 realization of disorder is reported in inset b together with the gaussian fit, $\bar{\lambda}/a$=10.18, σ_λ/a=0.827); and *iii)* Deviation from sinus curve (a blow up of the eigenvector, dots, is reported in the inset c) together with the best local sinus approximation,

full line. In the inset d) it is reported the residual of the eigenvector after the subtraction of the full line in the inset c).)

FIG. 2 Countour plot of the numerical joint distribution function $\mathcal{P}(\lambda, \lambda^{loc})$ obtained by the analysis of the modes at $\omega/\sqrt{K/M} \approx 0.031$. The countour plot has been obtained averagin 1,000 realization of disordered chains ($\Delta K/K = 0.3$) with N =1,000. The analysis has been performed with different choices for the weigth: a) P=1; b) $P = |\nabla e_p(i)|$; and c) $P = |\nabla e_p(i)|^2$. The $\mathcal{P}$-scale is logarithmic and the i-th line indicates the level at $10^{-i/4}\mathcal{P}_{max}$.

FIG. 3 Examples of $E(Q, \omega)$ vs Q at the indicated ω values calculated with the Dyson-Schmidt method [9] averaged over 50 realizations of disordered linear chains ($\Delta K/K = 0.6$) of length $N = 20,000$. Only the peak region is reported, and the full lines are the best lorentzian fits in this region.

FIG. 4 ω-dependence of the hwhm of the lorentzian fit to the $E(Q, \omega)$ reported in Fig. 3 (full dots) compared with the predictions from elastic constant fluctuations including (full line) or not including (dashed line) the localization effects.

FIG. 5 Examples of $S(Q, \omega)$ of simulated glassy Argon at the indicated Q values. In the inset the hwhm of the peaks of the $S(Q, \omega)$, calculated by a lorentzian fit in the peak region, (full dots) are reported vs Q together with the theoretical prediction from Eq. 6 (full line).